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**Final Report**

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**Soft X-ray Optics by Pulsed Laser Deposition**

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## Introduction

Of the growth techniques successfully used for fabrication of multilayered thin-film optics for soft x-rays Pulsed Laser Deposition (PLD) has been the least explored, because it is at an early stage of development. The main objective of this project was the fabrication and characterization of PLD-grown multilayered optics for soft x-rays with the goals of investigating the suitability of this technique, and lessening obstacles for its use, if possible. At the outset, the main difficulties perceived for PLD were (1) particulate emission and (2) lack of thickness uniformity. The first of these can be reduced in many cases by selecting an appropriate laser wavelength range, or by employing velocity selector filters, which are able to cut out the larger (and hence more sluggish) particles. Lack of film thickness uniformity due to the narrow forward peaking of the emission plume characteristic of the laser ablation process is a key issue affecting generalized applicability of PLD. The possibility of fabricating structures with large useful areas depends on this question, which was addressed in the course of this research project.

To allow fabrication of multilayers, a PLD system was prepared to allow automatic sequential deposition from multiple targets under computer control. Initially, the second or third harmonics of a Nd:YAG laser were used as the light source. Due to difficulties with beam uniformity and low UV output of this laser, an excimer laser was later incorporated as the main light source, operating at 248 nm (KrF excimer).

Software codes required for reflectivity calculations in the soft x-ray range were implemented and a database for optical "constants" in the EUV and x-ray region was updated. Reflectors for near-normal incidence for the  $\lambda = 125 \text{ \AA}$  region and for  $\lambda = 45.8 \text{ \AA}$  (i.e., near the carbon K edge) were designed. These structures were fabricated as test cases for the applicability of PLD.

## Results for Mo/Si multilayers

Mo/Si multilayers designed for  $125 \text{ \AA}$  operation were fabricated with UV light pulses with fluence at the targets of approximately  $3 \text{ J/cm}^2$  (power densities of  $3 \times 10^8 \text{ W/cm}^2$ ). The average deposition rates expected near the plume axis, from previous calibrations for Mo and Si, were  $0.08 \text{ \AA/pulse}$  and  $0.3 \text{ \AA/pulse}$ , respectively. Low-angle Bragg peaks were observed, but the results indicated in general poor

periodicity and a shift of the average bilayer spacing with respect to the intended one. This was attributed to angular deviation of the plume ejected from the targets, particularly Mo, that increased with dwell time. This would reduce layer thickness for the top layers, as well as affecting calibration runs. Individual Mo and Si films generally show a very smooth component, together with particulates, some of which are rather clearly due to rapid solidification of molten droplets ejected by the target. The particulates have sizes of order  $1\mu\text{m}$  and less, and are naturally undesirable since they would impair the reflectivity of any multilayer structure. While it was possible to produce Si films almost completely devoid of these particulates using 355 nm (third harmonic of the Nd:YAG), this had much to do with target surface condition. This is unfortunate because the minimum fluence required for target ablation produces rapid surface changes. The excimer laser offers the advantage of a smooth beam profile in comparison to the Nd:YAG beam. Also, Mo ablation with the excimer laser beam produces a plume with better characteristics in terms of particulate content. Unfortunately, it was found that Si ablation with the excimer laser (248 nm) beam generates a greater particulate density than the best previous results from the 355 nm beam. In addition, the particulate is largely in the form of irregularly shaped fragments, as opposed to the round frozen droplets observed mainly on samples produced with 355 nm light. This is attributed to the deeper penetration of the 248 nm light in Si, which can induce sub-surface heating and cause ablation in an explosive manner.

These results are disappointing, because the strong dependence on target surface condition, together with the related particle emission imply that PLD is not suitable for fabrication of Mo/Si multilayers which can meet the requirements of soft x-ray optics. It is possible that these difficulties can be partially overcome by using separate laser lines for each of the two targets. In this way, a more suitable wavelength could be used in each case. This approach requires simultaneous control of the two lasers, as well as of target motion. In the near future, we will be able to pursue this option. Naturally, real-time monitoring of film thickness would be very valuable, but this is far from trivial in PLD.

### **Results for C/Co multilayers**

The reflector design for  $45.8\text{ \AA}$  at normal incidence required C/Co multilayers with  $13.5$  and  $9.5\text{ \AA}$  layer thicknesses, respectively, with about 200 bilayers for

saturation. Film growth was carried out first from pyrolytic graphite and a Co sputtering target, using 355 nm laser pulses. Carbon ablation proceeds rather smoothly, without generation of many particles. Plume distribution appears to be wide compared to other materials observed. For Co, as for other highly reflective metals, yield is low. It was observed that plume deviation from normal occurs, although it is not as marked as in the case of Mo. Calibration lines for Co and C were deposited for a target-substrate distance of 4 cm, and their thicknesses measured with a Tolansky interferometer. Estimated average growth rates at a laser fluence of a few J/cm<sup>2</sup>, the minimum required for evaporation of Co, were 0.032 and 0.072 Å/pulse for Co and C, respectively. In principle, these rates should allow adequate thickness control, but this is of course spoiled by poor lateral thickness control. The similar yield of Co and C is advantageous for PLD growth via a source for which output power cannot be directly controlled "on the fly", such as our current Nd:YAG laser. Both Co and C films grown in individual preliminary runs are of good quality, with very smooth surfaces, as observed with optical microscope and SEM. In the case of Co there was some particulate due mainly to ejection of micron-sized liquid droplets.

C/Co multilayer samples were grown on Si wafers for XRD and Auger depth profiling analysis. While low-angle XRD scans (with a standard powder diffractometer) do show maxima near calculated Bragg peaks, these are poorly developed. This is not surprising because of (1) the relative importance of roughness for such thin layers, (2) the probable lack of in-plane layer uniformity inherent in the PLD process, and (3) the unsuitability of the diffractometer available for the task. On the other hand, the multilayer period estimated from the XRD results is within a few Å of the design value of 23 Å. Auger profiling of the samples indicated very good layering, considering the small layer thicknesses. This test is not conclusive however, because the ion beam mixes the materials and blurs the layering for such thin structures. This appears to be the case particularly after the first few layers from the top.

Growth of C/Co multilayers with the excimer laser was initiated by rate calibration at various fluences, starting with the lowest values that would produce noticeable plumes. Target-substrate distance was kept at 6 cm (relatively long for PLD) in order to favor film flatness. For carbon, relatively high fluences were required in order to obtain reasonable rates. At ~4 J/cm<sup>2</sup> fluence average growth was about 1.2 Å/sec (at the maximum laser repetition rate of 100 pulses/sec). The films appeared very smooth and with few particulates when observed with optical microscopy up to X 1000. Cobalt yield at 3 J/cm<sup>2</sup> fluence was about 0.2 Å/sec (also

at 100 Hz). The Co films appear also of excellent quality. This work will continue with deposition of additional C/Co multilayers and other C-spacer structures for further studies.

### Film coverage uniformity in PLD

Lack of beam uniformity is one of the key issues affecting generalized applicability of PLD. In particular, due to the tight thickness control required for multilayer soft x-ray optics, the possibility of fabricating structures with useful areas of any reasonable size depends on this issue to a greater degree than for most applications. While approaches such as beam scanning or substrate motion have been used before to enhance film thickness uniformity in PLD, these had not been systematized. Analysis completed in the course of this project can be useful in providing rules for fabrication of large-area films or multilayers by PLD with beam scanning. In addition, the details of a uniformity-enhancing scheme without beam scanning were worked out.<sup>1</sup> This utilizes annular light distributions at the target instead of scanning the beam into a circle. Means to generate annular light distributions onto the target for radially symmetric laser beams were designed. Two related patents were obtained from the U.S. Patent Office.<sup>2</sup> Target irradiation is caused to form an annular emission source instead of a spot. By calculating the resulting thickness profiles, an optimum radius  $s$  for the source is found, corresponding to a given power in the emission characteristic (modelled by a  $\cos^n \theta$  function with  $n > 1$ ) and a fixed target-substrate distance  $h$ . The results are illustrated in Figure 1. Calculations of optimal  $s/h$  ratios were performed for a wide range of values for  $n$ . Annular illumination of the target can be achieved for the case of a laser beam with radially symmetric profile by means of conic optics. The optical element devised for this purpose may be found useful in other applications. It is now covered by a separate patent.

Similar results in coverage uniformity can be obtained by scanning the laser beam on the target in circular paths with the same radii. In connection with this possibility, a novel type of laser scanner, suitable for application in PLD, was invented. A prototype was built and tested, and a technical disclosure was issued.<sup>3</sup> This scanner is an extension of a method known in the industry as "beam trepanning", which involves the motion of the focussing lens in a suitable path. This allows scanning in an elliptical path by very simple means. Elliptical paths are required in

order to produce a circular source pattern on a flat target inclined with respect to the incoming beam.

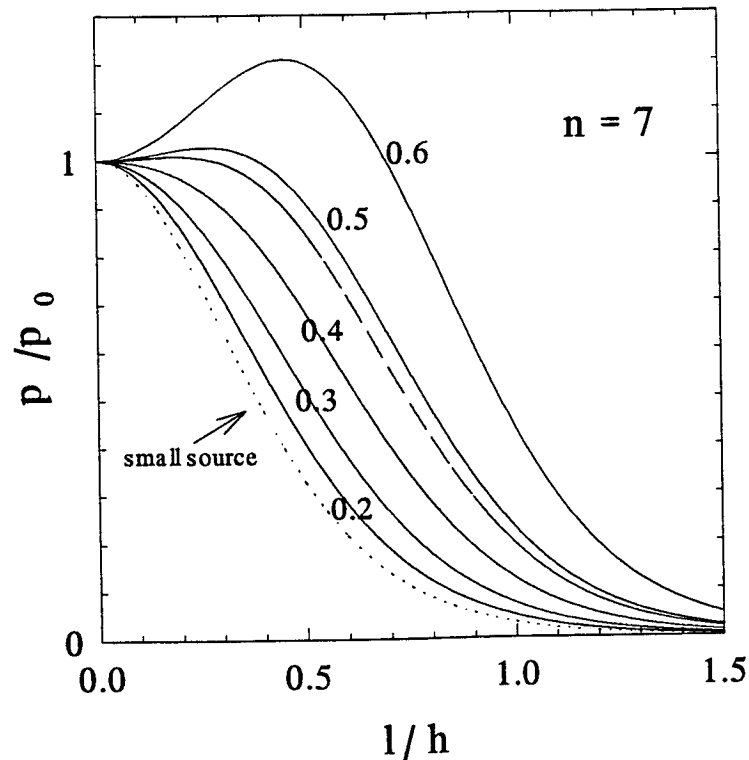


Figure 1. Calculated thickness profile, normalized to central (maximum) thickness, as a function of substrate radial parameter  $l/h$  (where  $l$  is the radial distance along the substrate, and  $h$  is the target-substrate distance) for target emission distribution of form  $\cos^7\theta$  and: small source (.....), thin annular sources with increasing radii,  $s/h = 0.2, 0.3, 0.4, 0.5, 0.6$  (————), optimum ratio,  $s/h = 0.475$  (- - - -). In this last case uniformity would be very good up to  $l/h = 0.3$ .

### Student participation

One graduate student completed his M.S. degree with a thesis based on this project. A second graduate student and an undergraduate have participated in aspects of the project. Two of the students presented their work in student technical meetings. Portions of this work were presented at state scientific meetings.

## Summary

Mo/Si and C/Co multilayers for soft x-ray optics were designed for spectral regions of interest in possible applications. Fabrication was effected by Pulsed Laser Deposition using Nd:YAG (355 nm) or excimer (248 nm) lasers in order to evaluate the suitability of this technique. Results for Mo/Si structures were not considered satisfactory due mainly to problems with particulate production and target surface modification during Si ablation. These problems may be alleviated by a two-wavelength approach, using separate lasers for each target. Results for C/Co multilayers are much more encouraging, since indication of good layering was observed for extremely thin layers. We expect to continue investigating this possibility. In order to compete with traditional PVD techniques, it is necessary to achieve film coverage uniformity over large enough areas. It was shown that this is feasible, and novel means of achieving it were devised.

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